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RADIOISOTOPE FUELED THERMOELECTRIC GENERATORS

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During the recent years a great attention has been paid to the design of low power self-contained sources utilizing the decay energy of radioisotopes . Since these power sources are extremely reliable and have a longer service life not requiring an additional charge they may be used to supply power to remote maintenance-free devices.

This report describes two radioisotope fueled thermoelectric generators designed in the USSR. The first power source utilizes Po-210 as a fuel, while the second one – Ce^{144}.

I. Radioisotope power source using Po^{210}

For recent years many papers have been published describing various radioisotope fueled thermoelectric generators delivering from 1 to 100 W. Lead selenides and tellurides are used as a thermoelectric material because of their higher figure of merit as compared with the well known materials. The figure of merit however is not the only factor indicative of thermoelectric material suitability. Application of high temperature alloys (although with the less figure of merit) allowed to increase the emitter temperature and so to save the weight of the device the efficiency being the same due to the greater temperature difference between hot and cold junctions.

A number of thermoelectric generators were designed
with the initial power up to 10 W , they had similar thermo-
electric conversion systems and differend only in a radiator
and in the way the thermopiles are pressed to the hot sur-
face. These instruments were used to determine equipment
capacity, electrical parameters and temperature distribu-
tion through the blocks by means of a radioisotope heat
source simulator.

Principle parameters. The preliminary investigation of
the device versions have showed that a design where a flat
capsule is snuggly fitted between hot surfaces of two
thermopiles the cold junctions of which are attached to the
radiator is a simplest one; this device has sufficiently low
heat loss.

The capsule has the shape of rectangular parallelepiped
of 60x60x13 mm. Manufacture technique and operating conditions
limit to some extent possible values of its parameters. For
example to make a capsule reliable at high pressure (up to
200 atmospheres) caused by helium produced in the decay its
surface temperature should not exceed $850^{o}C$. Temperature-resis-
tant rubber used for sealing the device limits the tempe-
rature of the radiator , which should not be more than $250^{o}C$.

A semiconductor section of the thermoelectric elements
was chosen 3 cm long; if the length were less the power would
fall off due to the higher commutation resistance in the
total thermal battery resistance and the insulation plate
resistance fraction in the thermopile resistance. On the
other hand, if the length 1 is more than 3 cm the weight of
the device builds up but its electric characteristics are
improved slightly. Silicon-germanium alloy used in this
device has an average figure of merit $Z = 4.10^{-4} 1/^{o}C$ and
a thermal conductivity factor $x = 1.5.10^{-2} cal/cm.sec.^{o}C$.

The results calculations made it possible to choose
the principle parameters, temperature characteristics and
efficiency of the device . The results are listed in
Table I.

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Conversion device. The silicon-germanium alloy was
used as a thermoelectric material of the conversion device
because of the following factors: a) high operation tem-
perature (above $1000^{\circ}C$); b) low vapor pressure at $T=1000^{\circ}C$;
c) high strength; d) resistivity against radiation; e) low
specific gravity. Each battery of thermogenerator comprises
16 thermoelements connected in series (8 thermoelements are
of 'p'-type and the other eight of 'n'-type), they are
screwed between two metal plates. The hot surface plate is
made of carbon steel and the fixing screws are of molyb-
denum. The cold surface plate and its fixing screws are
made of copper. Thermoelements screwed between steel and
copper plates are coupled by molybdenum wire contact
welded with the nickel foil 0.02 mm thick.

Both batteries are connected in series. In the instal-
lation 5 this connection is provided by a copper contact
insulated with ceramics; in installations 6 and 7 the housing
is used as a connecting conductor.

Installation housing. The housing of the installation 5
is of duraluminium and is cylindrical in shape, its bottom is
provided with a flange to form sealing with a copper top of
the housing. Two metal electrodes with ceramic insulation
are welded into the copper top. The shape of the copper top
flange is so chosen that the bottom can move 2-3 mm with
respect to the flange due to the top deformation. Gas-tight
coupling between the top and the housing is provided by
a sealing ring of temperature-resistant rubber.

Since the housing of installations 6 and 7 are of duralu-
minium throughout, they are more rigid and lighter, the
thermopiles are closer pressed to the radioisotope heat
source capsule. The thermobatteries of the installation 5 are
pressed to the radioisotope heat source capsule by means
of a disk steel springs fitted on bolts coupling two parts
of the housing. To provide good thermal contact between the
elements of the installation 7 the both parts of the hou-
sing are coupled by a clamp secured to the flanges. The
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low section of the housing has stiffening ribs. The construc-
tion of the top section permits bending of the bottom with
respect to the cylinder flange at a distance of 2 mm. The
holes are provided in the housing to remove air from the
interior and to fill it with the thermoinsulating material
"Perlit". To increase blackness factor the housing is
coated with a thin layer of hot proved paint with ξ =0.85.
Fig.I shows general view of the installation 7.

Electrical Simulator Used for Capacity Tests of Gene-
rator. The electrical simulator of the radioisotope heat sour-
ce is a graphite unit (size: 60 x 60 x 13 mm) with a molyb-
denum hairspring. Heater power was determined by connecting
a a.c. wattmeter into the heater circuit. The performances
of these systems were shown to be stable during 1000 hour
operation. The results are listed in Table 2. The efficiency
and electric power of thermogenerator 6 with the nuclide heat
source simulator were also determined under various tempe-
rature conditions. Fig.2 shows the test results.

Radioisotope Heat Source. Radioisotope heat source is a
rectangular parallelepiped; it is made of stainless steel.
Capsules containing Po^{210} are inserted in five cells.
The nickel capsule containing 1000-2000 curies of Po^{210} is
mounted within two stainless steel tubes for strength. Tubes
are provided with screw stoppers, welded during assembling.
Fig.3 shows a general view of the radioisotope heat source
and its components. The source thermal power was deter-
mined callorimetrically.

Tests of Thermoelectric Generator with Radioisotope
Heat Source . Generator 7 has undergone tests. The thermo-
generator with a nuclide heat source containing Po^{210} the
original thermal power of which equaled 244 W was set up
for 50 minutes, then it was connected to the meters. Accor-
ding to the bench test of generator 7, the following para-
meters wer continuously controlled: the voltage produced
by the thermogenerator across the load 0.34 ohm; the hot
skin temperature (two points); the radiating housing tempe-
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rature (two points); the gamma-radiation dose-rate at a
distance of 0.3 m from the source. The test results
are listed in Table 3.

Fig.4. presents volt-ampere characteristics taken perio-
dically (in five-seven days).

II. Electric Power Generating System Containing Ce^{144}

At present time the human life is hardly possible
without reliable and serviceable weather stations situated
both in inhabited regions and in remote almost inaccessible
ones, where there are no constant electric power sources.
The available power sources have definite limitations ham-
pering their usage and can not operate reliably and continous-
ly.

Electric power nuclide sources based on the thermo-
electric principle of converting the thermal power into
electricity are more reliable and have sufficiently high elect-
rical parameters.

The generator considered is a prototype of an electric
power source for automatic meteorological stations. The
possibilities of generators utilizing short lived beta-emit-
ting nuclides were determined by a test model with a heat
source containing Ce^{144} during controllable period (constant
output power) and uncontrollable one (output power falls off).

Performances. By the beginning of an uncontrollable pe-
riod the nuclide fueled generator contained 17500 curies of
Ce^{144}. Fig.5 presents the generator general view.

During an uncontrollable operation electric output is
5.0-5.6 W; voltage – 3.5 V ; overall efficiency 3.5 – 4%.
A thermopile based on solid d solutions of $Bi:Bi_2Te_3+Bi_2Se_3$
('n'-type conductivity) and $Bi_2Te_3 + Sb_2Te_3$ ('p'-type con-
ductivity) are used to convert the heat into electricity.
A special automatic device provides a predetermined power
level. The electric power is consumed by a storage battery
with a capacity of 12 A/hours, this battery supplies power
to a weather station, working in pulsed operation with 26 V
and 6 A. 318

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Radiation shielding consists of operating and shipping containers. The operating container shielding was designed to provide a dose rate not more than 1.0 r/hr at a distance of one meter from the generator surface. While the operating container inserted into the shipping one the dose rate reduces to the value not exceeding 10 mr/hr at a distance of one meter from the shipping container surface. The operating container has removable stiffening ribs, and the shipping one - fixed stiffening ribs. The generator may be used inserted into the operating container and into shipping one . Tungsten and lead are used as shielding material. The total weight of the generator with the operating container is 400 kg, the shipping container weight is 1200-1400 kg.

<u>Main Parameters and Engineering Design</u> . A necessary electric power of the generator is calculated assuming that the diurnal values of consumed energy and the delivered one are equal. The values of a necessary thermal power and an electric power for one disintegration assuming the gamma-absorption in the fuel, capsule and heat unit determine the radio-isotope activity.

While constructing the device a particular attention has been payed to ensure a required radiation shielding, a minimum temperature drop between a cold junction and an ambient air, a lighter weight and smaller overall dimensions. Several designs were considered having various tungsten heat block-to-lead shielding thickness ratio. The constructions differed in the way the thermopiles are replaced, in the thermal regulation (radiation, thermal shunt, insulation thermal conductivity); in the shape and material of the radiation shielding (spherical and cylindrical geometry); lead, tungsten and depleted uranium were considered as shielding materials.

The generator comprises: a) a heat and a radioisotope blocks; b) a thermopile; c) a thermal power controller; d) an arrangement for substitution of the thermopile; e) housing; f) a biological shielding.

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radioisotope
A block is fitted within a tungsten cylindrical heat
block with a diameter of 100 mm, its sides are 20 mm and
bottom is 40 mm thick. The cylinder is closed with a hemi-
spherical top fixed to the cylinder with a covering ring.

A isotope block is a double capsule made of stainless
steel XI8H9T-type. It consits of two coaxial cylinders
within which a vessel is inserted filled with a melted cerium
molybdate. Each cylinder is closed with a top, argon-arc
welded and is checked for air-tight sealing. The heat block
is mounted on the thermopile consisting of 97 thermoelements.
The thermopile is fixed in a special case and attached to the
bottom made of cylindrical copper plug. In order to maintain
nominal temperature conditions the generator is provided with
thermal control means including special thermal shields and
a control actuator with a reducing gear. A constant heat
flux through the thermopile (when isotope block power falls
off) is assured by reducing a radiating surface of the heat
block by moving the thermal shield. Fig.6 presents the ther-
mal power vs time. The spring thermal control actuator is
driven electrically. A thermal block, a thermopile and an
arrangement for substitution this thermopile are inserted
into a cylindrical housing with welded branches to mount a
case with a thermopile. A cap with a lead filler closes the
top end of the housing. Operation and shipping containers
provide a radiation shielding. Stiffening ribs of aluminium
alloy are used as a heat-removing system . This system
consists of 64 rectangular ribs fixed with a coupling bolt
on the operating container; these ribs provide a required
temperature drop between an ambient air and a thermopile
base. The conjugate surfaces of the dismountable elements
are highly finished and a sufficient specific pressure is
ensured at the point of contact.

Thermophysical Calculations. Cylindrical heat source
temperatures were calculated from a thermal conductivity
differential equation. Heat flux distribution over genera-
tor blocks were also determined. This distribution depenus
upon the thermopile operating conditions i.e. free running,
rated operation, and short circuit.

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The results are listed in Table 4.

4. Heat Generation Calculation

The heat generation in the unit, containing Ce^{144} is determined by beta-particles of $Ce^{144} + Pr^{144}$ and gamma-rays, absorbed in both isotope and heat blocks.

Space distribution of the specific heat generation due to beta-emission may be considered uniform throughout the capsule interior. Heat leakage due to bremsstrahlung are neg- lected since a bremsstrahlung transfered energy equals 1-2 per cent of the total energy; the most portion of this energy is absorbed in isotope and heat blocks. Space distri- bution of the specific heat generation due to gamma-radia- tion (it is of particular importance for nuclides of the Ce^{137} type) is defined by the following expression:

for gamma-radiation absorbed by the capsule material

$$Q_{\gamma_1}(\vec{r}) = \frac{3.7 \cdot 10^{10}}{4\pi} \, kq \sum_E E\, n(E)\, \mu_a(E) \int_V \frac{e^{-\mu(E)|\vec{r}-\vec{r}'|}}{(\vec{r}-\vec{r}')^2} \, B_a\left[\mu(E)|\vec{r}-\vec{r}'|\right] d\vec{r}'$$

Here $Q_\gamma(\vec{r})$ - heat generation per a volume unit close to the point with coordinate $\vec{r}$ (W/cm^2); $k = 1.6 \cdot 10^{-13}$ W.sec/Mev; q - material specific activity, curies/cm^3; n(E) - number of gamma-rays per one disintegration; V - capsule core volume; E - gamma-ray energy; μ (E) and μ_a (E) - gamma-ray attenuation coefficient and energy-absorption coefficient in the fuel respectively; $B_a[\mu$ (E) / $\vec{r} - \vec{r}'$ /] accummulated absorbed energy factor for gamma-radiation in the fuel.

Similar relations exist for gamma-rays absorbed by the capsule walls $Q_{\gamma_2}(\vec{r})$ and by the heat block $Q_{\gamma_3}(\vec{r})$.

The total heat generation is calculated by integrating the expressions for $Q_{\gamma_1}(\vec{r})$, $Q_{\gamma_2}(\vec{r})$ and $Q_{\gamma_3}(\vec{r})$ over drug, capsule and heat block volumes respectively. The heat generation during rated operation (controllable period end) was 135 W (17500 curies).

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Thermal power of the isotope block was measured by a special calorimeter, the obtained data agreed with the calculated ones.

Radiation Shielding Design. A multigroup method was used to design shielding, partial gamma-ray constants (the dose-rate given by the separate gamma line of the unshielded isotope) corresponding to gamma lines of $Ce^{144}+Pr^{144}$ was first determined. Bremsstrahlung was taken into account by replacing a continuous spectrum with a discrete one for each line of which a corresponding gamma-constant was determined. A series of partial gamma constants obtained was treated according to "competing lines" method that resulted in time saving.

The necessary heterogeneous shielding thickness was calculated from

$$\frac{[P]}{k} = q \sum k_{\gamma i} \; L_i \; B_i , \qquad (I)$$

where P — permissible dose rate; k — assumed safety factor ; q — drug specific activity ; k — partial gamma-constant; L_i — radiation function of the cylindrical source behind the shielding allowing for geometric factor, self-absorption in the source and attenuation in the shielding disregarding the scattered radiation (radiation function was calculated by an electronic computer); B_i — dose buildup factor allowing for multiple scattering of gamma-radiation within both the drug and capsule substance and the shielding.

The following expression was used for a dose buildup factor in a heterogeneous medium

$$B_i = \sum_{n=1}^{N} B_n \left(\sum_{k=1}^{n} \mu_{ik} d_k \right) - \sum_{n=2}^{N} B_n \left(\sum_{k=1}^{n-1} \mu_{ik} d_k \right) ,$$

$$(2)$$

where N — number of heterogenous shielding layers; μ_{ik} — attenuation coefficient of K-layer; d_k — K-layer thickness.

Expressions (I) and (2) were used to determine a dose rate on the shielding surface. It is necessary to notice that thicknesses calculated by this way agree with thickness values estimated according to Monte-Carlo method.

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Table 5 lists calculated values of two different shielding.

Thermoelectric generator and Voltage Transformer. A ther-
moelectric conversion device consists of 97 thermoelements
(5 mm x 5 mm) , 20 mm high. The thermoelements are made up of
solid solutions based on triple alloys $Sb_2Te_3+Bi_2Te_3$
and $Bi_2Te_3 + Bi_2Se_3$. A solder provides commutation allowing
to pperate in the temperature range of $300^{\circ}C$. To prevent the
thermoelements from oxidation at high temperatures hot jun-
ction are coated with a thin layer of a special enamel. The
total heat losses of the generator were calculated allo-
wing for the heat losses across the thermopile insulation
and the other blocks. The generator tests carried out by
means of an electrical simulator of the heat source under
normal operation conditions showed good agreement between
calculated and experimental data. Fig.7 presents generator
load characteristics obtained at controllable operation,
fug.8 – the same ones during the uncontrollable period after
50 days of operation under these conditions.

To recieve a d.c. voltage of 26 V this system com-
prises a transistorized dc-dc transformer. Energy storage
is accomplished by silver-zincum accumulators. The transformer
supplies current to the storage battery depending on the
operating conditions of the meteostation. The transformer
efficiency is 75 per cent; the accumulator efficiency is
75-80 per cent. The higher overall efficiency of 98 per cent
achieved with accumulators connected in series ,

Operation. The above generator with accumulators and a
voltage transformer is used as a power source at an automatic
weather station located in the middle part of the Soviet
Union. To minimuze a dose rate around the generator it is
slightly buried into the ground. The generator operation
was controlled by the output instruments which indicated
the principle parameters: voltage, electromotive force, current
All output parameters of the dc-dc transformer and accumu-
lators were automatically recorded.The operation recured
in every two hours. The output power at the end of control-
lable period was 5.4 W , voltage was 3.6 V.

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Fig. 9 shows variations of the dc-dc transformer input voltage (9-a), of the accumulator battery output voltage (9-b) and of the transformer efficiency for the whole cycle of operation . During an uncontrollable operation after two month period of 2 hr cycle the meteo station was transffered to 3 hr cycle. Climatic conditions were controlled during this experimental operation. The heat removal depended on these conditions. At present the system is in operation and if the power falls off the intervals between the operation should be made longer.

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<div align="right"><u>Table I</u></div>

1. Insulation thermal resistance, $\dfrac{{}^{o}C.sec}{cal}$ 12

2. Heat flow through insulation, W   49

3. Initial heat power of the capsule, W   320

4. Radiator surface, cm^2 900

5. Number of thermoelements (allowing
   for the external load voltage of 1.8 V)   32

6. Thermopile cross section, cm^2 0.8

7. Hot junction temperature, ${}^{o}C$ 760-775

8. Cold junction temperature, ${}^{o}C$ 300-330

9. Thermoelectric generator efficiency, %   3.5-3.9

10. Overall efficiency, %   3-3.3

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Table II

Nos.	Parameters	Installation number		
		5	6	7
1 : : 2	:	3 :	4 :	5
1.	Dimensions, mm			
	height	128	126	210
	diameter	195	196	190
2.	Weight (without the ra-dioisotope heat source capsule), kg	3.1	2.7	2.8
3.	Simulator heat power,Q, w			
	a) heat power equiva-lent of the radio-isotope heat source containing 10000 curies of Po^{210}	320	320	320
	b) heat power equvalent after three month of operation	205	205	205
4.	Electrical power of the installation, W:			
	a) when Q = 320 W	10.15	10.9	9.65
	b) when Q = 205 W	4.35	4.67	4.15
5.	Efficiency when Q=320 W,%			
	a) thermoelectric	3.75	4	3.56
	b) thermal	85	85	85
	c) overall	3.18	3.41	3.02
6.	Electromotive force when Q = 320 W, V	3.9	3.6	3.8
7.	Electromotive force when Q = 205 W, V	2.42	2.24	2.36
8.	Maximum temperature of the thermoelements hot plate when Q =320 W, $^{\circ}C$	861	840	817

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1 :	2	: 3 :	4 :	5

9. Radiator maximum
temperature when
Q = 320 W, $^{\circ}$C — 241, 250, 240

10. Maximum temperature
of the thermoelement
hot plate when
Q $\cong$ 205 W, $^{\circ}$C — 670, 650, 633

11. Radiator maximum
temperature when
Q = 205 W ,$^{\circ}$C — 190, 196, 195

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Table III

Nos.	Parameters	Operation period, hours			
		48	1000	1400	2000
1	Thermal power of the radioisotope heat source, W.	244	200	185	163
2	Electromotive force, V	2.85	2.3	2.09	1.88
3	Load voltage 0.34 ohms,V	1.4	1.14	1.04	0.92
4.	Power output, W	5.8	3.79	3.24	2.52
5	Installation efficiency, %	2.36	1.91	1.75	1.55
6	Thermoelectric generator hot plates temperature when operating at the load of 0.34 ohms, $^{\circ}C$	759 746	633 542	600 503	545 440
7	Radiator housing temperature, $^{\circ}C$	230 216	202 190	192 178	163 157
8	Activity of the radioisotope heat source using Po^{210}, curies	7700	6300	5320	5140

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Table IV

Operating conditions	Thermopile temperature drop	Heat flux via thermopile, W	Heat flux via the other blocks, W	Thermal efficiency
Free running	225	112.5	22.5	83.4
Rated operation	190	119	16	88
Short circuit	170	121	14	89.6

Note: Cold junction temperature assumed to be $0^{\circ}C$, the ambient temperature - $+20^{\circ}C$. Electric power of the thermopile is of 5.2 watts.

Table V

Direction	Version	Container type	
		Operating :	Shipping
Axial	1	Pb – 12 cm	Pb – 9.8 cm
	2	W-1 cm + Pb-10.I cm	Pb – 9.9 cm
Radial	1	Pb – 11.8 cm	Pb – 9.8 cm
	2	W-2 cm + Pb – 8.2 cm	Pb-9.9 cm

Note : Ce^{144} total activity of the source is 30000 curies; a dose rate is 1 r/h and 10 mr/h at a distance of 1 meter from the operating container and the shipping one respectively.

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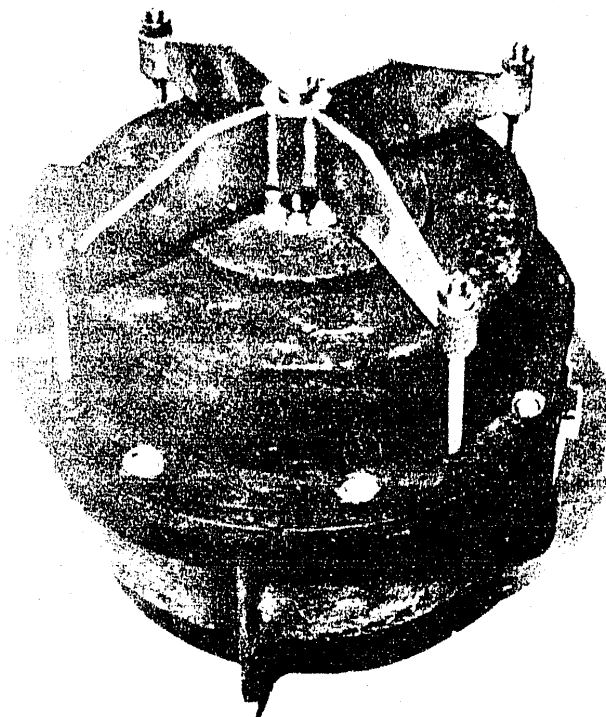
Fig.1. General view of the generator, containing Po^{210}
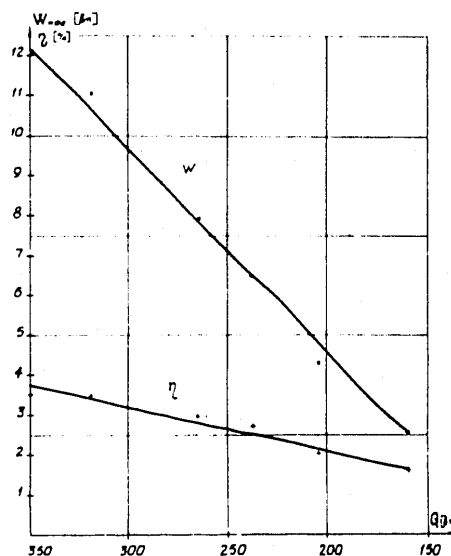
Fig.2. Electric power and efficiency of the installation
under various thermal conditions

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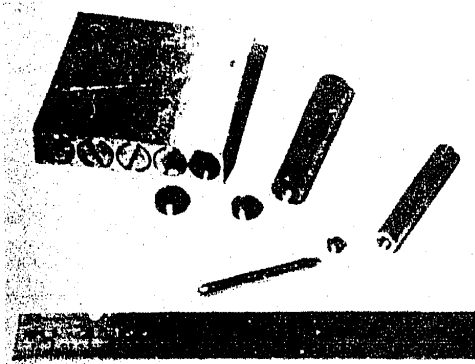
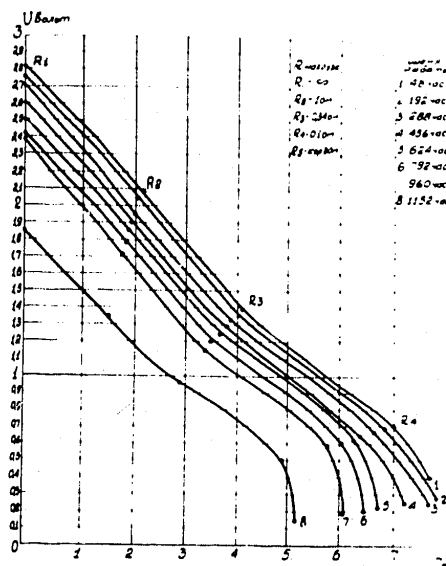Fig.3. Heat source capsule
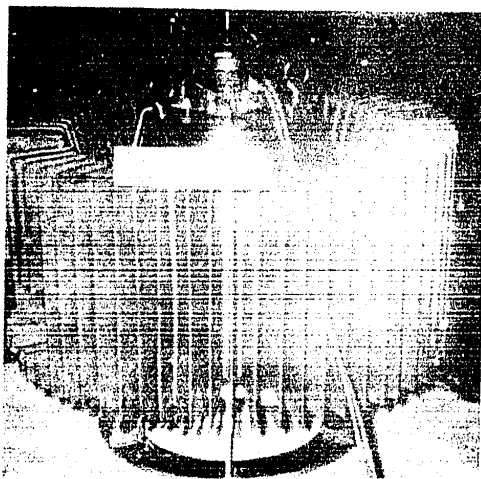and its components

Fig.4. Volt-ampere characteristics
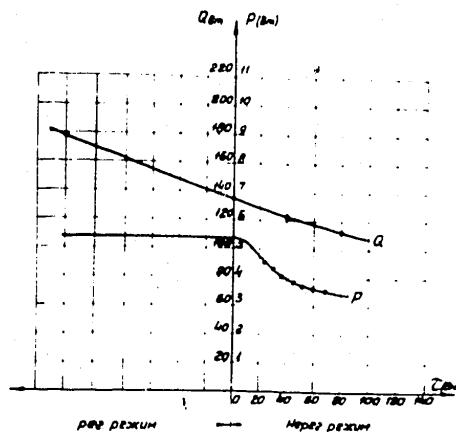

Fig.5. Generator general view
using Ce^{144}

Fig.6. Thermal (Q) and
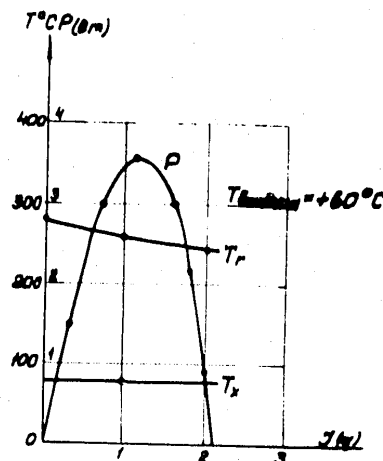output electric (P) power v.s.
operation time

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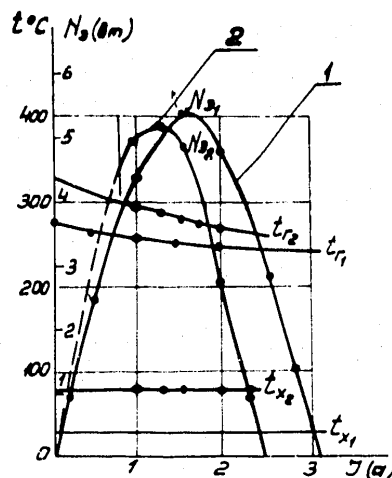
Fig. 7. Generator load characteristic
under controllable conditions
1 - ambient air temperature
+10°C
2 - ambient air temperature
+60°C

Fig.8 Generator load
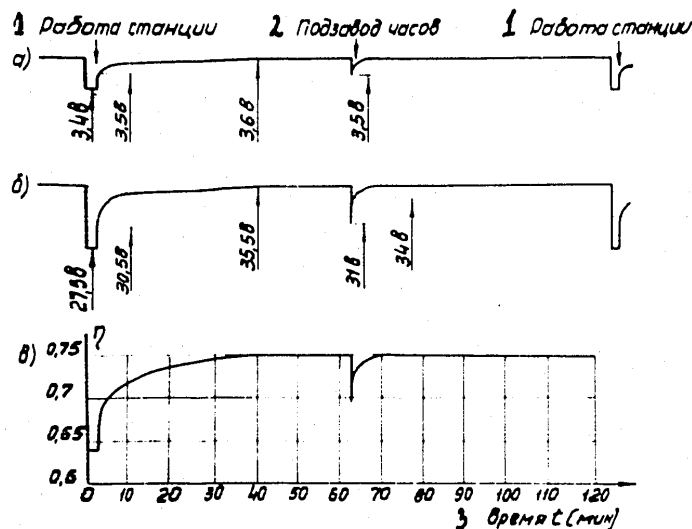characteristic under
uncontrollable conditions

Fig.9. The pattern of change of DC-DC transformer
input voltage(a), accumulator battery output
voltage(b) and transformer efficiency (c) for
one operating cycle of the weather station
1 - station operation; 2 - clock winding up
3 - time(min)

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